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PATENT SPECIFICATION

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COMPLETE SPECIFICATION

Improvements in the Production of Organic Oxygen-Containing Compounds

We, BADISCHE ANILIN- & SODA-FABRIK (I.G. FARBENINDUSTRIE AKTIENGESELLSCHAFT "IN AUFLOSUNG") a company recognised under German Law, of Ludwigshaven am Rhein, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The specification of Patent No. 644,665 which, though not published before the priority date of the present specification, has an earlier date, describes a process for the production of oxygen-containing organic compounds by adding on carbon monoxide and hydrogen to olefinic compounds (oxo reaction) while employing cobalt compounds, in particular cobalt carbonyls, as catalysts which are introduced into the reaction chamber in the form of volatile cobalt compounds. To achieve this, the carbon monoxide and hydrogen are passed through a zone containing metallic cobalt or a cobalt compound capable of forming volatile CO- and H-containing compounds under conditions favouring the formation of these volatile compounds.

We have now found that the said process may be carried out in a specially advantageous manner by producing the vaporous cobalt compounds by the continuous introduction of cobalt or cobalt compounds in a finely divided solid form into an otherwise empty vessel in countercurrent to the carbon monoxide or mixtures of carbon monoxide and hydrogen intended for the reaction and hereinafter called synthesis gas under increased pressure and at elevated temperature such as are necessary for the formation of the vaporous colbalt compounds.

The amount of cobalt or cobalt compound, as for example cobalt oxide. hydroxide or carbonate, and the speed of flow are so chosen that the synthesis gas advantageously contains from 0.2 to 5 grams, in particular 0.5 to 1.5 grams, of Co in the form of gaseous cobalt compound per cubic metre (at normal temperature and pressure). It is preferable to supply the cobalt or cobalt compounds to the whole of the synthesis gas; it is also possible, however, to charge a partial current of synthesis gas with the gaseous cobalt compound and then dilute this with more synthesis gas, or also to charge only carbon monoxide or a partial current thereof and later to mix this with the hydrogen. In all cases it is preferable to carry out the production of the gaseous cobalt compounds under increased pressure and at about the temperature at which the synthesis gas is caused to react with the olefine in the oxo reaction chamber.

The cobalt material may be introduced into the gas current in various ways. One of these possibilities is shown diagrammatically in the accomponying drawing.

Referring to the drawing, a constant amount of pulverulent or finely grained cobalt metal or cobalt oxide is continuously withdrawn from a pressure-tight reservoir A by means of a conveyor worm B operating at constant speed and introduced into the upper end of a high pressure pipe C which is heated to about 140° C. by heating devices D_1 and D_2. The synthesis gas employed for the reaction, or part of the same, is led upwardly through this tube. The cobalt material entering the gas current is converted to volatile cobalt compounds, mainly carbonyl and hydrocarbonyl, which are entrained by the current of synthesis gas and led through a heated pipe connection E into the reaction vessel F. Under the catalytic influence of the entrained cobalt compounds, the reaction between the synthesis gas and the olefine introduced through the pipe G takes place in the reaction vessel F with the formation

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of normally liquid, for the most part monomeric aldehydes. By means of a pressure equalising pipe (not shown), care is taken that the same pressure of, for example, 250 atmospheres prevails in the whole apparatus.

The invention is not limited to the employment of a conveyor worm. Any other charging device may be used instead of the same. For example it is also possible to allow a constant amount of cobalt material to trickle into the pipe C from a reservoir above the same simply by means of a slide or regulatable valve.

In order that it may not be necessary to interrupt the process when the cobalt material in the reservoir A is used up, a second chamber may be arranged above the reservoir A which can be independently released from pressure or placed under pressure and from which cobalt material introduced into the same may be emptied through a valve into the reservoir A. When the cobalt material in A is almost used up, the upper container is opened, freshly filled, placed under pressure and the introduced amount emptied into A. It is also possible to work with two reservoirs in parallel.

The following example, given with reference to the drawing, will further illustrate this invention but the invention is not restricted to this example.

EXAMPLE.

8 kilograms of powdered commercial cobalt oxide are charged into the reservoir A. By means of the worm B, running at a speed of about 1 revolution per minute, 13 grams of colbalt oxide (corresponding to 9 grams of cobalt metal) are introduced per hour into the upper end of the high pressure pipe C which is 10 metres long and has an internal diameter of 24 millimetres. Through the pipe C which is heated to 140° C., there are led per hour 10 cubic metres (NTP) of a gas mixture consisting of 55% of carbon monoxide and 45% of hydrogen under 250 atmospheres. The cobalt oxide falling into the gas current is completely converted into $Co(CO)_4H$, $Co(CO)_4$ and H_2O, so that the gas current leaving the upper end of the tube contains about 900 milligrams of Co per cubic metre (NTP) in the form of volatile carbonyl compounds. The cobalt-containing gas mixture is led through the heated connecting pipe E into a second high pressure pipe having a diameter of 90 millimetres and a length of 10 metres, which is heated to 150° C. and into which about 15 litres per hour of liquid isobutylene is pumped at the top. The hourly output is 10 to 11 litres of a liquid product consisting to the extent of 90% of monomeric isovaleraldehyde. The yield remains the same throughout an uninterruped operation time of about 25 days. The cobalt oxide in the reservoir A is then used up and the reservoir must be refilled.

What we claim is:—

1. A process for the production of organic oxygen-containing compounds by adding on carbon monoxide and hydrogen to olefinic compounds while employing as catalysts vaporous cobalt compounds, in particular cobalt carbonyls, which are introduced into the reaction chamber in vopour phase, wherein the vaporous cobalt compounds are produced by the continuous introduction of cobalt or cobalt compounds in a finely divided solid form into an otherwise empty vessel in counter-current to the flowing carbon monoxide or mixture of carbon monoxide and hydrogen intended for the reaction under increased pressure and at elevated temperature such as are necessary for the formation of the voporous cobalt compounds.

2. A process as claimed in Claim 1 wherein the finely divided solid cobalt-containing material is introduced by means of a conveyor worm into the space through which the gas containing carbon monoxide is flowing.

3. The process for the production of oxygen-containing compounds substantially as described in the foregoing example.

4. Oxygen-containing compounds when obtained by the process claimed in any of Claims 1 to 3.

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1 SHEET
This drawing is a reproduction of the Original on a reduced scale.